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G Boyle
For: H. F. Gray, Supervisor
Laboratory Records Dept.
ORNL

June 3, 1952

Dr. David H. Gurinsky, Head
Division of Metallurgy
Brookhaven National Laboratory
Associated Universities, Inc.
Upton, Long Island, New York

Dear Dave:

Please find enclosed the memorandum on "Fuel Elements for ORNL Graphite Reactor." I hope that this will supply all the information which you will need for the handbook. If not, please feel free to call upon us for any further information.

Best regards,

ORIGINAL SIGNED BY
E. J. BOYLE

E. J. Boyle
Assistant Director
Metallurgy Division

EJB:BB

Distribution: [redacted]

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[redacted] person is prohibited.

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X-426 (Revised 1-52)

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CENTRAL FILES NUMBER
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DATE: JUNE 3, 1952

SUBJECT: FUEL ELEMENTS FOR ORNL GRAPHITE REACTOR

TO: Dr. David H. Gurinsky, Head, Div. of Metallurgy
Brookhaven National Laboratory, Assoc. Universities, Inc.
Upton, L. I., New York

FROM: E. J. Boyle and C. D. Cagle

DISTRIBUTION: ~~1A--Dr. David H. Gurinsky~~
~~2A--M. E. Ramsey, X-10~~
~~3A--J. A. Cox, X-10~~
~~4A--C. D. Cagle, X-10~~
5A--E. J. Boyle, X-10

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~~SECURITY INFORMATION~~

This document consists of 13 pages.
No. 2 of 5 copies, Series A

# UNBONDED, ALUMINUM-JACKETED SLUGS

## Canning Procedure

These slugs were prepared by drawing 2S aluminum cans onto uranium slugs by inserting the slug into the can and forcing the assembly through a sizing die. Two types of slugs, with the following specifications, were employed:

1. Resistance-welded

   Aluminum Can

   Material--2S, impact-extruded
   Length--4.375"
   I.D.--1.116"
   Wall thickness--0.017" to 0.020"

   Uranium

   Material--Gamma-extruded
   Length--4.000" ± 0.010"
   Diameter--1.100" ± 0.002"
   Chamfer on bottom end--0.010" radius

2. Arc-welded

   Aluminum Can

   Material--2S, impact-extruded
   Length--4.375"
   I.D.--1.116"
   Wall thickness--0.035"

   Uranium

   Same as for resistance-welded

The first type of slugs, which had the end closure made by resistance welding of an aluminum end cap to the aluminum can, were used only during the first year of pile operation (1943-1944) and were replaced by the arc-welded slugs. In the case of the arc-welded slugs, the end closure was carried out by forcing an aluminum cap of size 1.102" diameter, 0.060" thick into the open end of the aluminum can after the sizing-die operation, trimming the excess metal from the can wall and lip around the end cap, and, finally, sealing with a Heliarc weld at the junction between can and cap.

## Testing Procedures

Five different procedures were used for testing of the slugs prepared as described above. These were as follows:

1. **Hydrogen Test**

This test involved holding the slugs for ten hours at 300°C in a hydrogen atmosphere of 15.5 p.s.i. The basis for the test was that any leaks in the slug would admit hydrogen with consequent formation of uranium hydride, which would cause swelling of the jacket. The test was rejected when it was found that hydrogen would diffuse through the jacket of sound slugs.

2. **Deflection Test**

Slugs were subjected to a pressure of 175 p.s.i. of nitrogen after heating to 300°C for one hour. The heating was intended to exert tension on the weld to expose any incipient leaks. After exposure to nitrogen, the slugs were measured with a micro-gauge deflection instrument to detect distortion (which would be caused by nitrogen's entering a leak) of the bottom of the jacket to within 0.001". The test was abandoned when it was found to be nonreproducible.

3. **Bubble Test**

It was planned to subject the slugs to a pressure of 175 p.s.i. of nitrogen and then submerge in kerosene to observe any bubbles which might be released from leaks. The method was abandoned when it was found that the sudden release of pressure in the pressure chamber fogged the observation port and also because of the long-time delay in bubble formation.

4. **Weight (Borst) Test**

This test was based on the fact that uranium reacts rapidly with oxygen of the air at elevated temperatures. A slug held at elevated temperatures, therefore, would gain weight as a result of oxidation of the uranium. Weighing the slug before and after a heating would reflect any change in weight.

A charge of slugs was given an initial heating period of 24 hours at 290°C $\pm$ 10°C to remove any volatile matter adhering to the jackets. After cooling, each slug was given an initial weighing to a tenth of a milligram and then placed back in the oven for a ten-day heating period at 290°C $\pm$ 10°C.

On completion of the heating period, the slugs were reweighed to a tenth of a milligram and classified as accepted, retested, or rejected material. A statistical method of accepting and rejecting the slugs was developed to compensate for the inaccuracies of the balances and errors made by the operators. The accepted slugs were then stored in the vault for pile use. Retest slugs were stored until enough accumulated to provide for a retest charge, which was tested in the same manner as new material. Rejected slugs were stripped and returned for rejacketing. Theoretically, holes as small as $10^{-6}$ inches in radius can be detected by this method.

The testing of slugs by the weighing method was completed on February 10, 1945, with the following results:

| | | |
|---|---|---|
| Number of slugs tested | 104,058 | |
| Number of slugs accepted | 100,118 | 96.21% |
| Number of slugs rejected | 3,940 | 3.79% |

The causes for the rejection of the 3,940 slugs were broken down as follows:

| | | |
|---|---|---|
| Jackets swollen or burst by oxide formation or diffusion(?) | 1,255 | 31.9% |
| Jackets swollen by gas formation | 611 | 15.5% |
| Excessive gain in weight | 481 | 12.2% |
| Pitted, scarred, and concave jackets | 547 | 13.8% |
| Rejection by visual inspection of weld | 1,046 | 26.6% |

5. Modified-Pressure Bubble Test

This method for the testing of slugs originated at Argonne National Laboratory and was further developed at ORNL. The slugs, after a visual inspection, were heated to $300^{\circ}C \pm 10^{\circ}C$ for a period of five days. Upon removal from the oven, any that showed signs of swelling or rupture were discarded. All of the others were loaded into an autoclave with the welded end up. The slugs were exposed to 300-p.s.i. helium pressure for 16 to 18 hours. Immediately after the release of the helium pressure, the slugs were placed end up under acetone, and the end was covered with a gas collector. After remaining under the acetone for five hours, all slugs which showed the release of gas bubbles were rejected. Holes as small as $10^{-6}$ to $10^{-7}$ inches in radius should have been detected by this method.

The results of testing of a large number of slugs by this method were as follows:

| | | |
|---|---|---|
| Slugs tested | 33,629 | |
| Slugs accepted | 33,176 | 98.65% |
| Slugs rejected | 453 | 1.35% |

The causes for the rejection of the 453 slugs can be broken down as follows:

| | | |
|---|---|---|
| Jackets swollen or ruptured during heating period | 217 | 47.9% |
| Pitted or scarred jackets | 25 | 5.5% |
| Slugs leaking gas | 211 | 46.6% |

Upon completion of the foregoing tests on Hanford-prepared slugs, approximately 7,000 slugs that had originally passed the weight test were retested by this method with the following results:

| | | |
|---|---|---|
| Number of slugs tested | 5,120 | |
| Number of slugs accepted | 5,094 | 99.49% |
| Number of slugs rejected | 26 | 0.51% |

The 26 unaccepted slugs were rejected because of leaking gas from the jackets and not because of swelling during heating or because of scarred jackets.

## Operational Experience

Since the start of reactor operation in November, 1943, there have been 99 known primary* slug failures with a total of 127 slugs involved. This amounts to a percentage failure of 0.056%. A study of the collected data makes it apparent that, given a full reactor loading of new unbonded slugs at one time, the ruptures that might occur with time would begin in the center rows of the reactor and proceed from there to the periphery. Since only the neutron flux and temperature of the reactor are distributed in this manner, it is fair to assume that one or both of these influence the cause or causes of the ruptures. From the known cases of rupture positions, the following important distribution can be made:

| | |
|---|---|
| Slug ruptures east of center line | 5 |
| Slug ruptures west of center line | 46 |

The distribution of all ruptures whose position was known is shown in Figure 1. The ruptures extend in an eastward direction only a distance of six and one-half slugs or 26 inches but extend in a westward direction a distance of 27 slugs or 189 inches. As may be seen in Figure 2, the maximum temperature occurs considerably west of the center line. Neutron flux is symmetric in the east and west directions from the center line, as shown in Figure 3. It seems reasonable, then, to assume that temperature is the most important factor in slug ruptures.

There are two possible rupture mechanisms which would be very temperature-dependent. These are as follows:

1. Oxidation

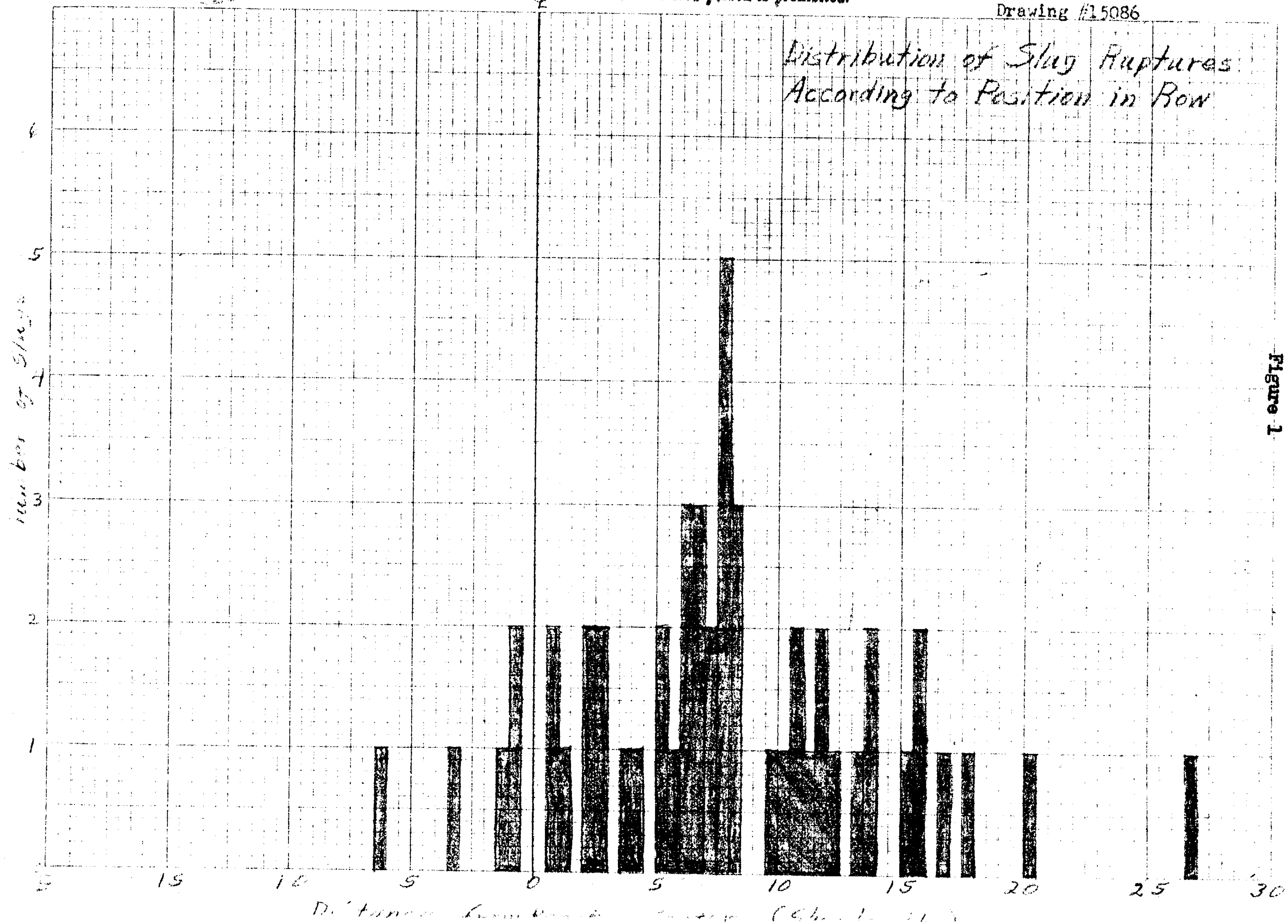
If a tiny hole is present in a slug jacket when it is charged into the reactor, it is inevitable that rupture will occur eventually if the slug is in a sufficiently high-temperature zone for a sufficiently long time.



*A primary failure is one in which no adjacent slugs are involved. Secondary failures are those in which a primary failure has caused partial stoppage of air cooling with consequent overheating of adjacent slugs.

Drawing #15086

Figure 1



Drawing #15084

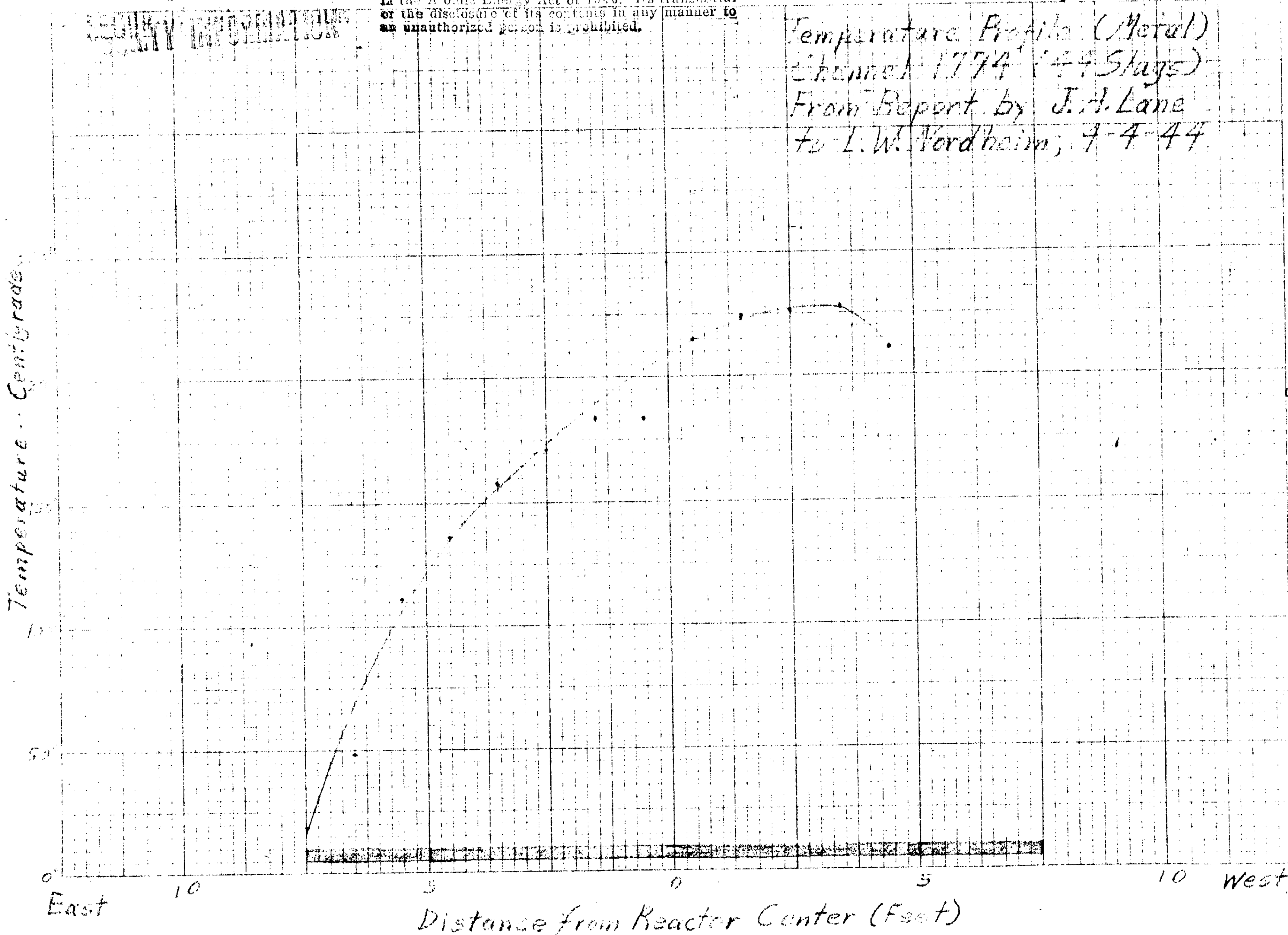



Figure 2

Figure 3

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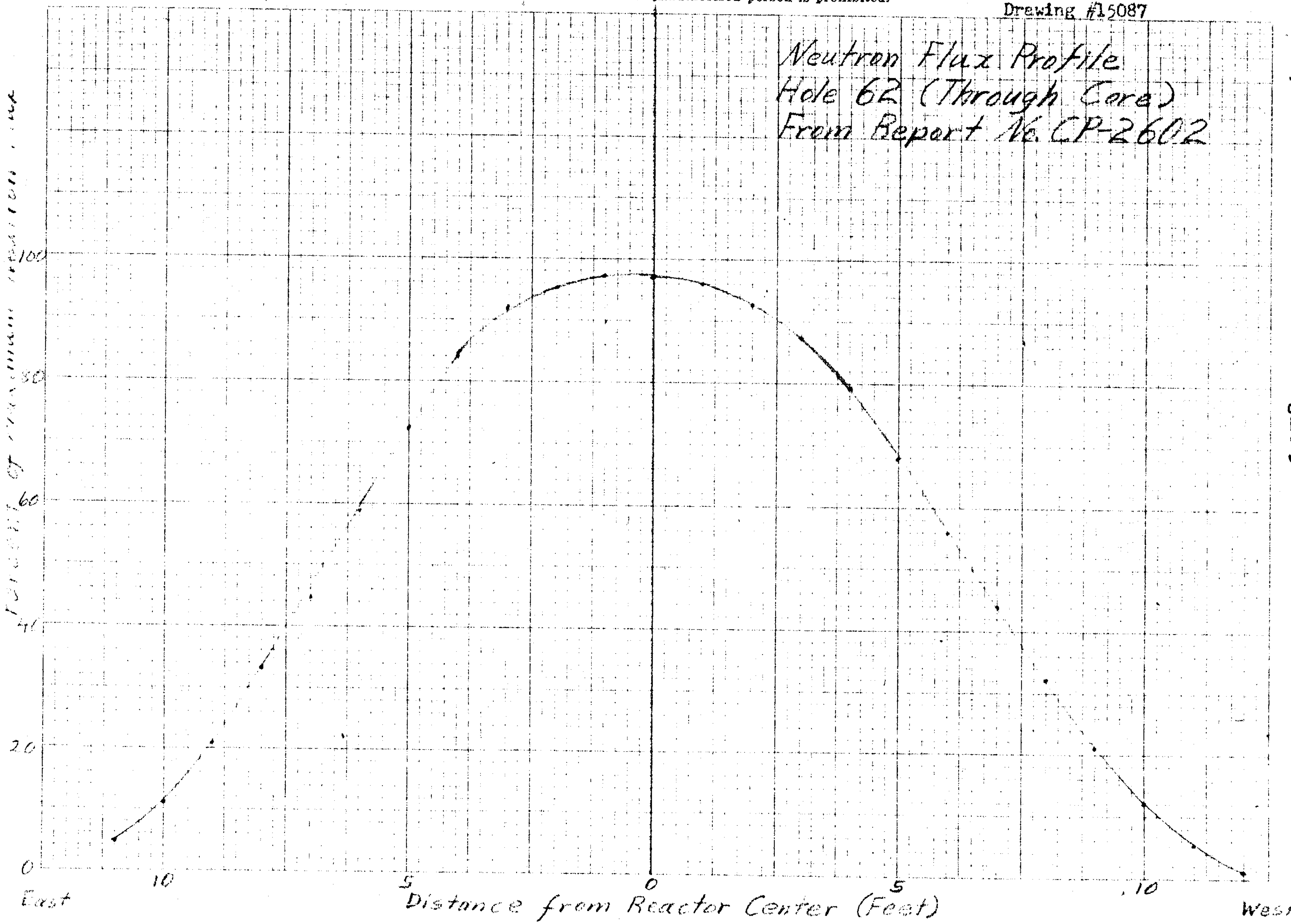

Figure 4, from BR-223, May 13, 1943, shows that the oxidation rate for uranium at 250°C is about 27 times that for 100°C. Assuming that there are two slugs with the same size holes in their jackets and that the same amount of oxide buildup is necessary for rupture of the aluminum cans, it would take a time period 27 times as long for the slug at 100°C to rupture as that for the one at 250°C. This may explain the delayed rupture of slugs which have been in the lower-temperature zones of the reactor for three to seven years.

2. Diffusion between the aluminum jacket and uranium slug has been shown to take place at an appreciable rate at 250°C. It will, of course, occur at lower temperatures at slower rates. Out-of-pile heating tests have shown conclusively that rupture can result from this diffusion. Visual examination of the inner-jacket surfaces and uranium-slug surfaces of stripped slugs that had been discharged from the reactor showed evidence of diffusion. Since diffusion is accelerated by an increase in temperature, this mechanism would seem to fit the temperature pattern for ruptures.

Figure 4

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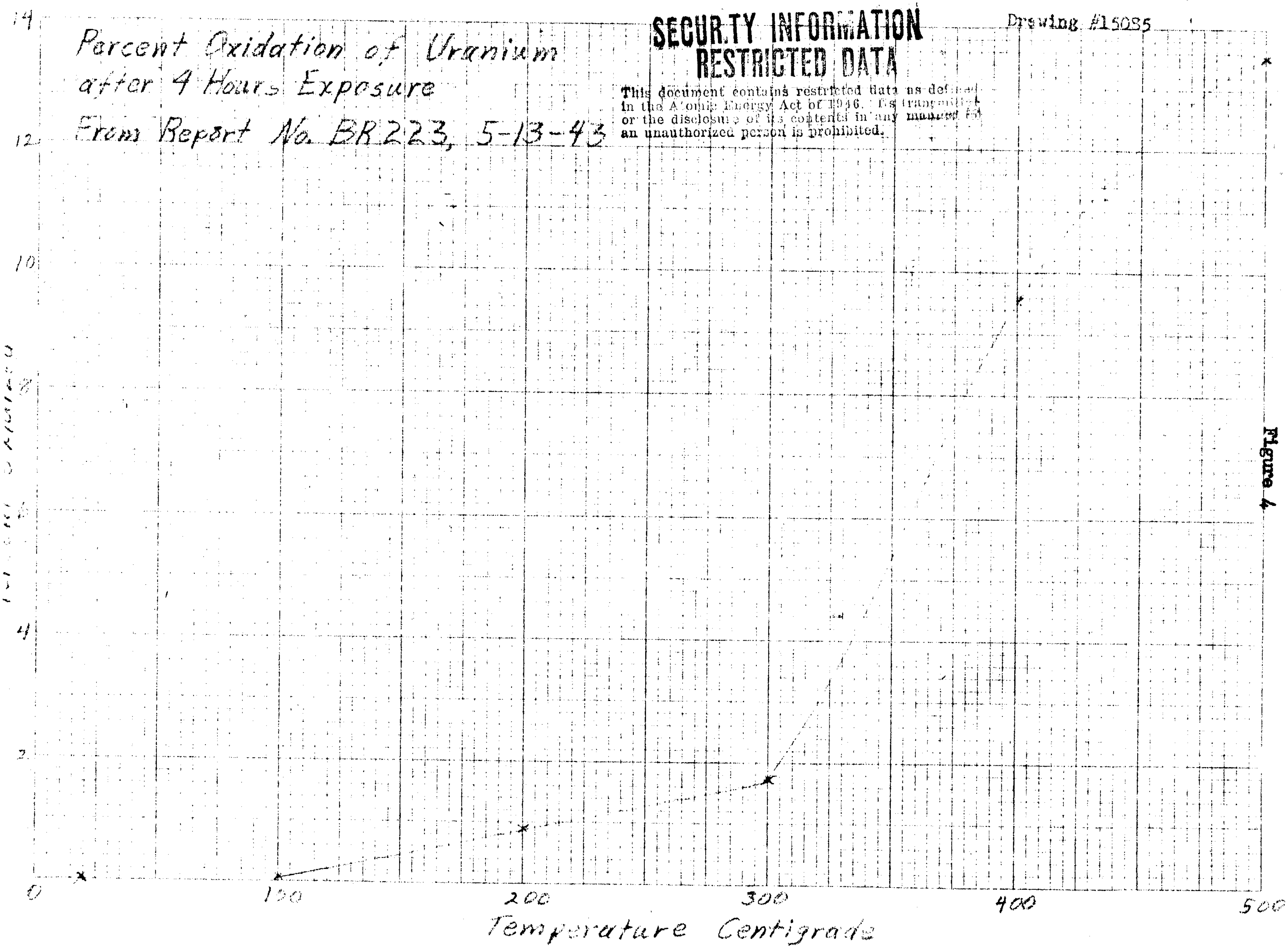

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## ALPHA CANNING OF ALUMINUM-SILICON BONDED SLUGS

### Canning Procedure

The decision to adopt aluminum-silicon bonded slugs as the standard fuel units for the ORNL air-cooled, graphite reactor was based on the following reasons:

1. Since the majority of the ruptures appeared to result from leaks in the can welds, aluminum-silicon bonded slugs are superior, since the closure consists of the double protection of a weld in the can plus the aluminum-silicon bonding layer.

2. The interdiffusion of aluminum and uranium, known to be a major cause of failure in unbonded slugs, is practically eliminated in bonded slugs by the presence of the diffusion barrier, $U(Al, Si)_3$, which is formed on the uranium-slug surface during immersion in molten aluminum-silicon alloy.

It had been planned originally to employ the standard production technique used at Hanford, i.e., the triple-dip process, to produce the required slugs for the ORNL reactor. It was found that a large percentage (about 20%) of these slugs exhibited diffusion blistering during the standard culling test of holding at 400°C for seven days. Since it was found that this diffusion blistering was intimately associated with high residual tin content, it was decided to adopt the alpha-canning process in order to avoid the tin bath of the triple-dip process.

Alpha canning of slugs for the ORNL X-10 graphite pile is is carried out in the Chemical Department at the Y-12 Plant. The dimensional specifications for the slugs are as follows:

Uranium Slug

Diameter -- 1.102" $^{+0.000"}_{-0.003"}$

Length -- 4.000" $^{+0.005"}_{-0.000"}$

End radius -- 0.060" $^{+0.040"}_{-0.000"}$

Material -- alpha-rolled

Aluminum Can

I.D. -- 1.121" $^{+0.000"}_{-0.003"}$

Wall thickness -- 0.033" $^{+0.000"}_{-0.003"}$

Bottom thickness -- 0.051" ± 0.005"

Length -- 4-21/32" ± 1/64"

Material -- 2S, deep-drawn



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Canned Slug

Length -- 4.120" ± 0.006"
Bond layer thickness -- 0.004" to 0.0065"

The steps in the canning procedure are as follows:

a. Beta Heat Treatment. This step, considered a preliminary step prior to canning, is carried out by immersing slugs in a salt bath at 720 to 740°C for a period of three minutes. The salt used is Houghton's Liquid Heat No. 980. Slugs machined to size prior to the salt-bath treatment were used, because they had already been machined for the triple-dip process. If the Y-12 Canning Group were starting anew, the salt-bath treatment would be carried out prior to machining, because some pitting of the slugs occurs during the salt-bath treatment.

b. Duplex Bath. After chemical cleaning (50% nitric acid) following the salt-bath treatment, the slugs are immersed in a duplex bath consisting of a ten-inch-thick lead layer at the bottom and a 15-inch-thick aluminum-silicon layer at the top. A cover of Eutector 190 flux (manufactured by Eutectic Welding Company) is kept on top of the aluminum-silicon layer. The slugs are held for 35 seconds in the lead layer at 600°C and then raised into the aluminum-silicon layer and held for five seconds at 600°C.

c. Aluminum-Silicon Dip Bath. Slugs are transferred from the duplex bath to the dip bath (11.2% silicon), where they are held for 13 seconds at 600°C.

d. Aluminum-Silicon Canning Bath. After the slugs have been wet with aluminum-silicon in the dip bath, they are transferred to the canning bath, where they are inserted into a preheated aluminum can which is inside of a steel sleeve. Just prior to insertion, the aluminum can is filled with aluminum-silicon by lowering the mouth below the surface of the bath. The final operations are to insert a preheated aluminum cap, prewet with aluminum-silicon, into the end of the can and quench the entire assembly into water.

e. Welding. After cutting end caps to proper length, a weld is made at the end cap-can interface with inert-gas, shielded-arc welding.

Testing Procedures

Two of the standard Hanford inspection tests--i.e., visual inspection and autoclaving--were used. The autoclave test consists of holding the slugs for 40 hours under 125-p.s.i. steam pressure. A third Hanford test, the frost test, was eliminated. Finally, a test consisting of holding the slugs for one week at 400°C was employed to determine whether diffusion between the uranium core and the aluminum can would occur.

Approximately 20,000 alpha-canned slugs have been subjected to the heat-treatment culling test of holding for one week at 400°C. Of these, 25 have shown blisters or have ruptured. In every case examined, the blisters or ruptures appear to have arisen from oxidation which was initiated by leaks in the welds or in the end caps. None could be attributed to diffusion.

Operational Experience

The recharging of the ORNL X-10 graphite pile is now underway, and approximately 20,000 bonded slugs have been charged. There has been too little service time to draw any conclusions about the behavior of these slugs under operating conditions. It may be noted, however, that some experience has been gained with the use of aluminum-silicon bonded slugs in general in the X-10 pile. One complete channel of the pile was charged in December, 1949, with triple-dip canned slugs prepared at Hanford. This channel has been operated with a reduced air flow so that the maximum surface temperature of the slugs is 330°C as compared with the usual 250°C maximum. To date, no ruptures or visual distortion of these slugs has occurred.

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3. E. A. Smith, HW 9401--Operating Process for Canning Four-Inch Slugs

4. E. J. Boyle, ORNL 1275--Application of Alpha Canning to Preparation of Slugs for ORNL X-10 Graphite Pile